

PATENT COOPERATION TREATY

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INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

REC'D 25 MAY 2005

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Applicant's or agent's file reference LU6096		FOR FURTHER ACTION		See Form PCT/PEA/416
International application No. PCT/EP2004/003452		International filing date (day/month/year) 01.04.2004	Priority date (day/month/year) 03.04.2003	
International Patent Classification (IPC) or national classification and IPC C08F210/16, C08F200, C08F2/38				
Applicant BASELL POLYOLEFINE GMBH				
<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 6 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input checked="" type="checkbox"/> sent to the applicant and to the International Bureau) a total of 2 sheets, as follows:</p> <p><input checked="" type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).</p> <p><input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.</p> <p>b. <input type="checkbox"/> (sent to the International Bureau only) a total of (Indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>				
<p>4. This report contains indications relating to the following items:</p> <p><input checked="" type="checkbox"/> Box No. I Basis of the opinion</p> <p><input type="checkbox"/> Box No. II Priority</p> <p><input type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p><input type="checkbox"/> Box No. IV Lack of unity of invention</p> <p><input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p><input type="checkbox"/> Box No. VI Certain documents cited</p> <p><input checked="" type="checkbox"/> Box No. VII Certain defects in the international application</p> <p><input type="checkbox"/> Box No. VIII Certain observations on the international application</p>				
Date of submission of the demand 27.10.2004		Date of completion of this report 27.05.2005		
Name and mailing address of the international preliminary examining authority:  European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016		Authorized Officer Kaumann, E Telephone No. +31 70 340-3640		



INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No.
PCT/EP2004/003452

Box No. I Basis of the report

1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
 - ☐ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
 - ☐ international search (under Rules 12.3 and 23.1(b))
 - ☐ publication of the international application (under Rule 12.4)
 - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report):*

Description, Pages

1-42

as originally filed

Claims, Numbers

1-12

received on 19.04.2005 with letter of 14.04.2005

- ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing
3. ☐ The amendments have resulted in the cancellation of:
 - ☐ the description, pages
 - ☐ the claims, Nos.
 - ☐ the drawings, sheets/figs
 - ☐ the sequence listing (*specify*):
 - ☐ any table(s) related to sequence listing (*specify*):
 4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
 - ☐ the description, pages
 - ☐ the claims, Nos.
 - ☐ the drawings, sheets/figs
 - ☐ the sequence listing (*specify*):
 - ☐ any table(s) related to sequence listing (*specify*):

* If item 4 applies, some or all of these sheets may be marked "superseded."

**INTERNATIONAL PRELIMINARY REPORT
ON PATENTABILITY**

International application No.
PCT/EP2004/003452

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims	1-12
	No: Claims	
Inventive step (IS)	Yes: Claims	1-12
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-12
	No: Claims	

2. Citations and explanations (Rule 70.7):

see separate sheet

Box No. VII Certain defects in the international application

The following defects in the form or contents of the international application have been noted:

see separate sheet

Re Item V

**Reasoned statement with regard to novelty, inventive step or industrial applicability;
citations and explanations supporting such statement**

1. Subject-matter

Subject-matter of the present application is a start-up process for the preparation of polyolefins in a gas phase fluidized bed reactor using a metallocene catalyst, which is specified in claim 11 as bis(1-methyl-3-butyl-Cp)ZrCl₂.

The start-up phase has a duration of from 30 minutes to 30 hours.

The melt flow rate of the obtained product (190°C, 2,16 kg) is less than 4 g/10 min but during start-up, a product having a melt flow rate higher than 4g/10 min is produced.

According to claim 7, the melt flow rate is adjusted by the hydrogen concentration.

The technical problem underlying the subject-matter of the present claims appears to provide a trouble-free smooth start-up process avoiding the formation of fine particles and chunks.

2. Prior Art

D1 = WO 03/022888 discloses a fluidized bed gas phase polymerization process, wherein the amount of off grade product is reduced during the transition from a first product to a second product, wherein the melt index is adjusted by changing the hydrogen concentration (see page 8, lines 16 - 26). It is mentioned that the second product's melt index can be lower than the first product's melt index.

No numerical values are disclosed for the melt index.

D1 does not mention a start up process.

D2 = US 5,077,358 discloses a fluidized bed gas phase polymerization process comprising a Start Up process using a Ziegler Netta catalyst. In comparative example 3, table 2, the melt flow rate varies with the time and drops from 7,2 to 4,1, but not lower. Later, it rises again. No formation of agglomerates is observed.

D3 = WO 02/50127 discloses a fluidized bed gas phase polymerization start up process, wherein the melt index is below 10, preferably below 1 (see page 38, lines 19 - 22).

The hydrogen partial pressure is kept constant throughout the whole process (see page 39, line 27 to page 40, line 3).

D4 = EP 0 571 826 (cited in the application) discloses a fluidized bed gas phase polymerization process for the preparation of ethylene copolymers using a Ziegler-Natta catalyst. The reaction temperature is defined using the formulae of the present application. There is no disclosure for the start up or a variation of the melt flow rate.

3. Article 33(2) PCT (Novelty)

It appears that novelty has to be acknowledged to the subject-matter of the present **claims 1 - 12** regarding the available prior art.

4. Article 33(3) PCT (Inventive Step)

D1 does not relate to a start up process but discloses the transition from a first product to a second product, wherein the melt index is adjusted by changing the hydrogen concentration. The catalyst is not specified but can be chosen from a list comprising Ziegler-Natta catalysts and metallocene catalysts.

D2 relates to a fluidized bed gas phase polymerization start up process using a Ziegler Natta catalyst. A melt flow rate of lower than 4,1 is not disclosed.

D3 relates to a fluidized bed gas phase polymerization start up process, wherein only a melt index below 10, preferably below 1 for the obtained products is disclosed.

The process according to **D3** uses a mixture of two catalysts. There is no suggestion that the melt flow rate could be changed as a feature of the start up process.

No disclosure or suggestion can be derived from the available prior art which would render the claimed process obvious.

Therefore, an inventive step can be acknowledged.

5. Article 33(4) PCT (Industrial Applicability)

Since the gas phase polymerization of olefins is a very important industrial process, industrial applicability of the subject-matter of present **claims 1 - 12** is evident.

Re Item VII

Certain defects in the international application

**INTERNATIONAL PRELIMINARY
REPORT ON PATENTABILITY
(SEPARATE SHEET)**

International application No.

PCT/EP2004/003452

Present claims 2 and 10 appear to be redundant.

We claim:

1. A process for starting-up an olefin polymerization reaction in a gas-phase fluidized-bed reactor using a catalyst comprising a metallocene to produce a polyolefin having a melt flow rate at 2.16 kg and 190°C in accordance with ISO 1133 of less than 4 g/10 min, wherein a polyolefin having an increased melt flow rate of above 4 g/10 min is produced during a transitional period during a start-up phase having a duration of from 30 minutes to 30 hours, in particular from 2 hours to 20 hours.
2. A process as claimed in claim 1, wherein the start-up phase has a duration of from 30 minutes to 30 hours, in particular from 2 hours to 20 hours.
3. A process as claimed in either of the preceding claims, wherein the melt flow rate during the start-up phase is initially above 4.5 g/10 min and is continually decreased to the value below 4 g/10 min.
4. The process as claimed in any of the preceding claims, wherein the temperature is increased by at least 1°C compared to the temperature in long-term operation, at least prior to the start-up phase.
5. A process as claimed in claim 4, wherein the temperature is increased by from 1.5 to 4°C.
6. A process as claimed in claim 4 or 5, wherein the temperature during long-term operation of the reactor is in a range bounded by an upper limit given by equation I

$$T_{RH} = 170 + \frac{6d'}{0.84 - d'} \quad (I)$$

and a lower limit given by equation II

$$T_{RN} = 173 + \frac{7.3d'}{0.837 - d'} \quad (II)$$

where the variables have the following meanings:

T_{RH} maximum reaction temperature in °C
 T_{RN} minimum reaction temperature in °C

d' value of the density d of the polymer to be produced.

7. A process as claimed in any of the preceding claims, wherein the melt flow rate is regulated via the hydrogen concentration in the reactor.

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8. A process as claimed in any of claims 1 to 6, wherein the melt flow rate is regulated via the monomer partial pressure in the reactor.

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9. A process as claimed in any of the preceding claims, wherein the polyolefin is a homopolymer or copolymer of ethylene.

10. A process as claimed in any of the preceding claims, wherein the organic transition metal compound is a metallocene.

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11. A process as claimed in claim 8, wherein the metallocene is bis(1-methyl-3-butylcyclopentadienyl)zirconium dichloride or bisindenylzirconium dichloride.

12. A process as claimed in any of the preceding claims, wherein an alkylaluminoxane is used as activating compound.

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